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OBSERVATIONS OF WATER STABLE ISOTOPES TO STUDY CHANGES IN LARGE SCALE ATMOSPHERIC TRANSPORT

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ABSTRACT

Water vapor is the most important green house gas in the Earth Atmosphere. The stratospheric concentration is dominated by the amount of water crossing the tropopause in the tropics as part of the large scale Brewer-Dobson circulation and by the oxidation of methane. Increases in stratospheric water could increase the frequency of polar stratospheric clouds and change the radiative balance of the atmosphere. An increase in the water vapor concentration over the last decades has been observed, but can only be partially explained by the increase in methane and indicates a change in the amount of water transported into the stratosphere. Water isotopes are particularly suited to help to quantify this contribution, and detect changes in the upper troposphere and lower stratosphere (UTLS). Detailed understanding of UTLS transport pathways is key to predicting the future of the ozone layer. We propose long term measurements of stable water isotopes from the SOFIA Upper Deck using a near-infrared tunable diode laser sensor.

INTRODUCTION

Water vapor is the most important greenhouse gas. Detailed understanding of the water cycle and water transport into the stratosphere is important for prediction of future climate change. Both radiative forcing and chemistry are affected by water vapor in the stratosphere. Stratospheric water vapor concentrations have an effect on both the production of OH radicals and the formation of polar stratospheric clouds, which modulate polar ozone destruction. In the stratosphere H_2O is also created by methane oxidation. Remote sensing and in-situ measurements indicate a trend of increasing water vapor concentrations in the stratosphere in recent decades that can not be fully accounted for by the increased methane concentration. However, recently it has been noted that this trend has reversed in satellite data [Randel et al., 2004]. Nevertheless, we do not fully understand the reason for these changes.

The main entry point into the stratosphere for water vapor is in the tropics. Deep convection carries humidity into the tropical tropopause layer (TTL, extending from 14-19km [Sherwood and Dessler, 2001]) and seldom penetrates the stratosphere directly. There are currently several different hypotheses for the dehydration of air entering the stratosphere. One set of hypotheses is centered on convective-scale motions involving overshooting cloud turrets and ice particle sedimentation [convective dehydration, e.g., Sherwood and Dessler 2000]. The second concept postulates that air detrains from convection at the bottom of the TTL, but with abundant water compared to the stratosphere. While slowly ascending, large-scale quasi-horizontal motions through regions where the cold-point temperatures are anomalously low, such as the “cold trap” of the Western Pacific, lead to the formation and subsequent sedimentation of

particles that dehydrate the air [Holton and Gettelman, 2001]. Testing of all these hypotheses requires improved observations, and an improved understanding of transport processes, in the TTL.

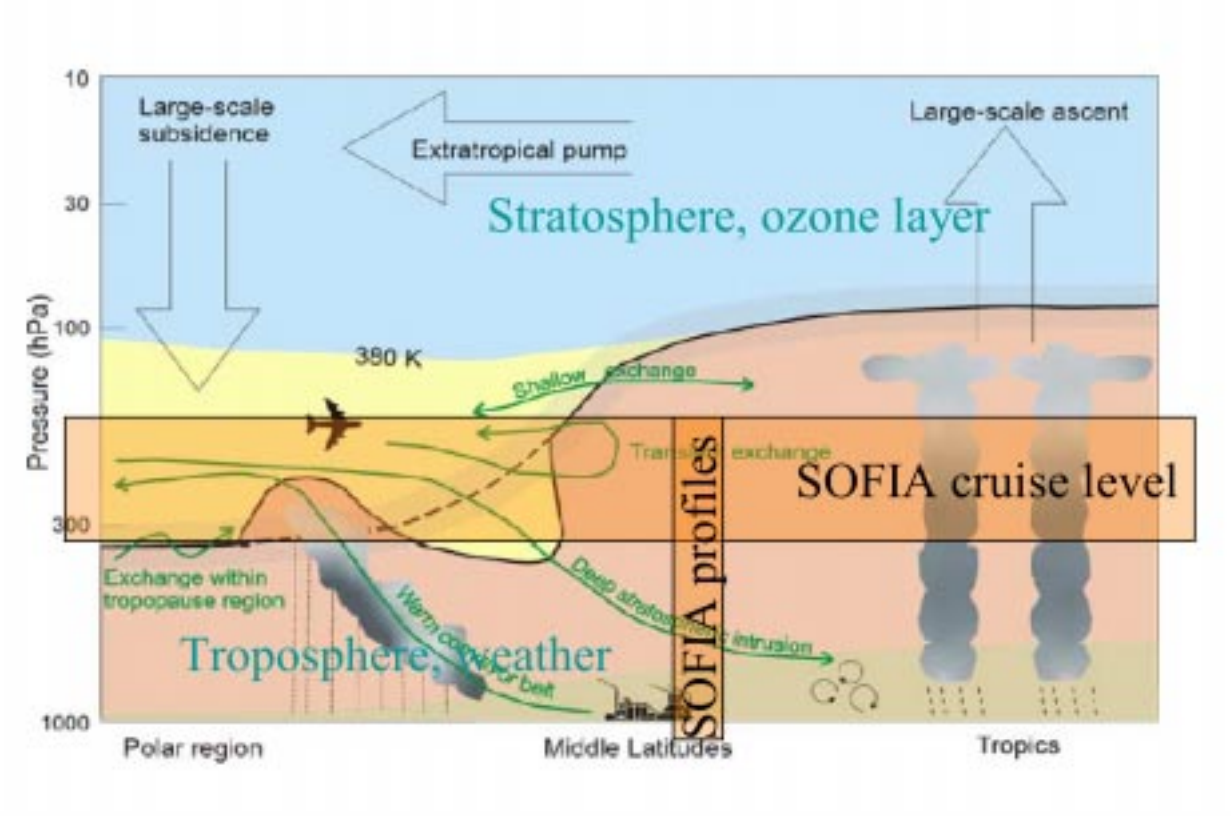

Figure 1: Schematic of stratosphere-troposphere exchange and approximate SOFIA coverage. Water isotope ratios contain key information to assess the transport path of water into the stratosphere. The different pathways of stratosphere-troposphere exchange are indicated by the green arrows. From [*Stohl et al.*, 2003]

Few observations at mid-latitudes have been made so far. Due to the unique signature added by the temperature history of an airmass, Zahn et al [1998] found that water vapor in the mid-latitude lowermost stratosphere had the composition of tropospheric air at low latitudes and supported the efficient adiabatic transport of air across the tropopause [Holton et al., 1995]. More recently, Wang suggested a way to transport pathway for water vapor into the stratosphere over mid-latitude convective systems [Wang, 2003]. These airmasses would again have a distinct isotopic signature and regular sampling of the tropopause region would allow us to estimate the budget of this process.

In winter at high latitudes stratospheric dehydration by falling ice particles and subsequent sublimation in the upper troposphere will show a typical signature in HDO and $H_2{}^{18}O$ [Zahn et al., 1998]. Slightly elevated HDO and $H_2{}^{18}O$ above the typical sampling air temperature dependent line would be expected from these falling particles.

Currently, NASA satellites are not capable of measuring water isotopes with the necessary resolution in the critical tropopause region. Measurements aboard SOFIA offer a unique opportunity to fill this gap and give us insight into crucial atmospheric water circulation.

INSTRUMENT DESCRIPTION

We describe an instrument to measure water isotope ratios in the gas phase, which performed engineering test flights on the NASA DC-8 in Dryden in May 2004 and would be very well suited for integration on SOFIA Upper Deck. The device flew on a total of 4 flights covering altitudes from 1000 ft above ground to 41,000 ft. The device performed very well during these flights. Figure 2 shows a typical in-flight spectrum obtained during the last flight. There are two lines of the main isotope $^{1}H_{2}{}^{16}O$, and one each of $^{1}H^{16}O^{2}H$, $^{1}H_{2}{}^{17}O$, and $^{1}H_{2}{}^{18}O$ in our laser scan.

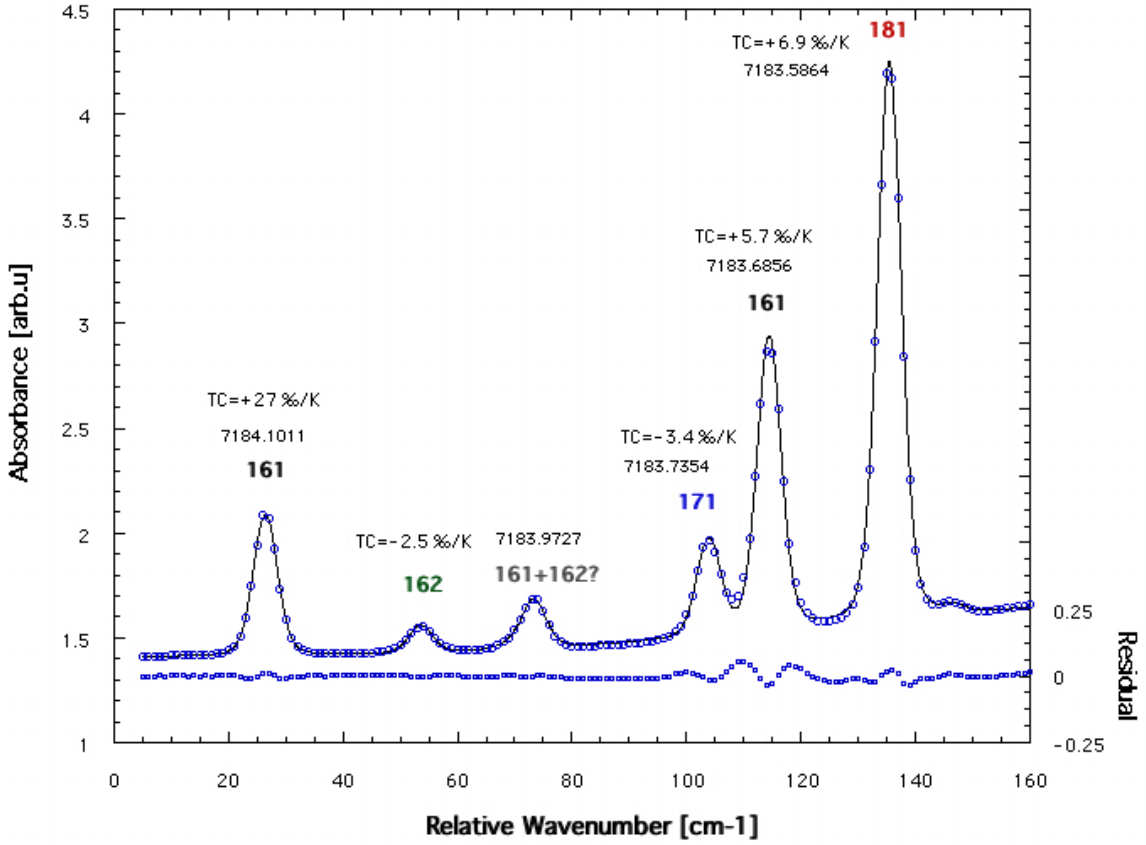

Figure 2 Typical in-flight spectrum and fit obtained during the DC-8 flights in May 2004. Each spectral feature is labeled by the temperature dependence of the line, the center frequency and the isotopologue number.

Our device is based on the technique of Optical Feedback Cavity Enhanced Spectroscopy (OFCEAS) which was developed at the University of Grenoble by Dr. Romanini and coworkers [Morville et al., 2003]. The most serious problem of 'conventional' cavity ring-down or cavity enhanced absorption spectroscopy using CW lasers is the difficulty of injecting laser light into a spectrally very narrow high-finesse cavity. Traditionally, the solution has been to use an extremely fast feed-back loop to lock the laser to the cavity transmission, or to rely on accidental overlap, possibly aided by mechanical vibrations. The OFCEAS technique, in contrast exploits optical feedback from a V-shaped high finesse (F > 104) cavity (cavity physical size L=~1m) in order to increase the light injection level into the spectrally narrow cavity modes (10 kHz typically). During a laser frequency scan (induced by a current ramp) the cavity returns feedback to the laser only when one of its longitudinal modes is excited. This produces a temporary frequency self-locking of the laser emission to that mode, in addition to a laser line width

narrowing which increases the mode injection. This way, the cavity output peak intensity may be recorded for several successive modes in a laser scan. The absorption lines of a gas present are superimposed on the cavity transmission peak intensities. These absorption lines appear enhanced as if they were observed as the output of a multi-pass cell with an effective length given by ~F*L or about 10km, resulting in an extremely high detection sensitivity. We record the cavity transmission mode-by-mode, which means that the spectral points are equally spaced and calibrated (separated by the cavity FSR~150MHz).

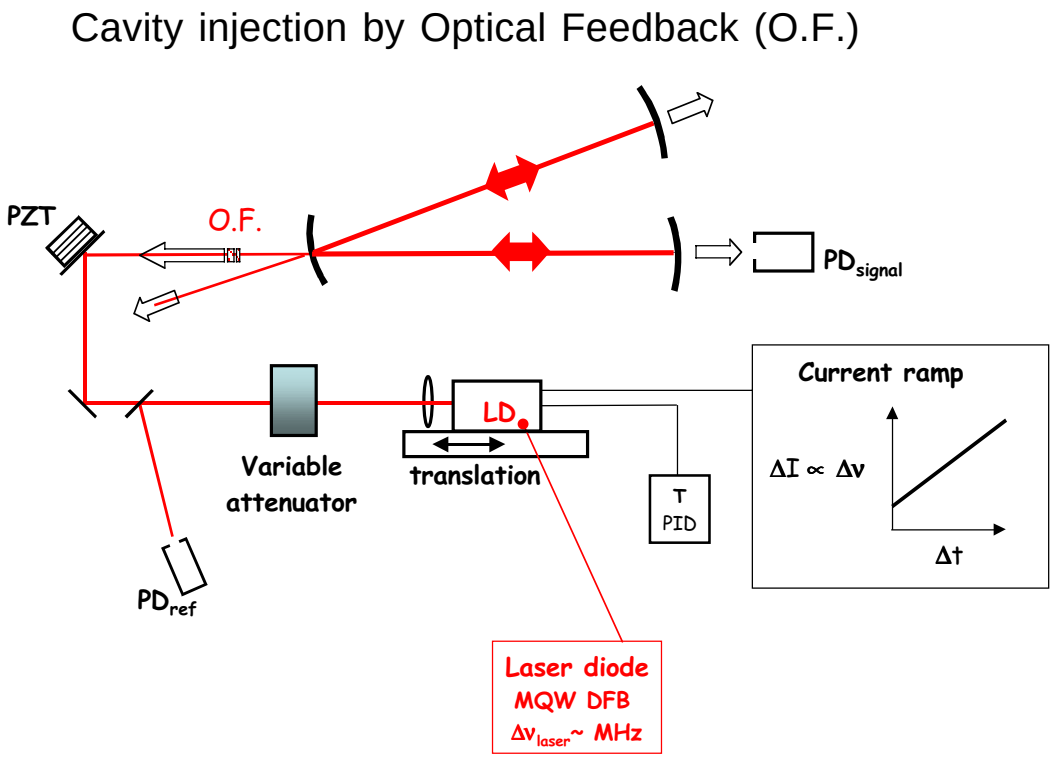

Figure 3 Optical Feedback Cavity Enhanced Absorption Spectroscopy (OFCEAS) layout. PZT: piezo-electrically mounted mirror; PD: photodiodes;

There is thus negligible noise on the frequency scale. The setup is very simple and uses few optical components (

Figure 3): A temperature-controlled DFB laser, collimated by a standard aspheric lens; A V-shaped optical cavity terminated by 3 high-reflectivity mirrors; 2 photodiodes: One at cavity output, the other monitoring the laser power picked-off from a beam splitter; A variable attenuator placed at the laser output controls the feedback level from the cavity to the laser; A piezo-electrically mounted mirror controlled by an electronic servo-loop which stabilizes the laser-cavity distance in real time.

Relatively short sections of the isotope record acquired during the engineering test flights in May 2004 may be used to assess the preliminary level of precision. During level flight at an altitude of 41,000 ft, assuming the inlet conditions did not change, the isotope signal may be used to calculate a short term (~20 min) precision level of 1.4‰ for 10 second integration. In exactly the same way, we determined precision levels of 10‰ and 15‰ for ^{17}O and ^{2}H, respectively. These values were later confirmed by laboratory measurements.

SUMMARY

Even if SOFIA will not sample the tropical tropopause region in the first years of operation, extended observations of water isotope ratios in the mid- and high latitude tropopause region will significantly improve our understanding of the aridity of the stratosphere. Insight into hitherto unexplained magnitude of stratospheric water trends will be possible considering the long term deployment plans for SOFIA. Detection of future changes in transport pathways into the stratosphere due to climate change will be uniquely feasible from the SOFIA platform. Long term trends due to climate change will likely be manifested by tropopause temperature changes, and should be reflected in the isotopic composition of stratospheric air. Tunable diode laser based measurements of the isotope ratios will offer unprecedented horizontal and vertical resolution.

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REFERENCES

Holton, J.R., and A. Gettelman, Horizontal transport and the dehydration of the stratosphere, *Geophys. Res. Lett.*, *28*, 2799–2802, 2001.

Holton, J.R., P.H. Haynes, M.E. McIntyre, A.R. Douglass, R.B. Rood, and L. Pfister, Stratosphere-Troposphere Exchange, *Rev. Geophys.*, *33*, 403-439, 1995.

Morville, J., D. Romanini, and M. Chenevier, Dispositif à laser couplé à une cavité par rétroaction optique pour la détection de traces de gaz. / Laser device coupled to a cavity by optical feedback for detecting gas traces, Patent FR2830617, (11 April 2003)

Randel, W., F. Wu, S. Oltmans, K. Rosenlof, and G. Nedoluha, Interannual changes in stratospheric water vapor, and correlations with tropical tropopause temperatures, *J. Atmos. Sci.*, *in press*, 2004.

Sherwood, S.C., and A.E. Dessler, A model for transport across the tropical tropopause, *J. Atmos. Sci.*, *58*, 765–779, 2001.

Stohl, A., P. Bonasoni, P. Cristofanelli, W. Collins, J. Feichter, A.Frank, C. Forster, E. Gerasopoulos, H. Gäggeler, P. James, T. Kentarchos, H. Kromp-Kolb, B. Krüger, C. Land, J. Meloen, A. Papayannis, A. Priller, P. Seibert, M. Sprenger, G.J. Roelofs, H.E. Scheel, C. Schnabel, P. Siegmund, L. Tobler, T. Trickl, H. Wernli, V. Wirth, P. Zanis, and C. Zerefos, Stratosphere-troposphere exchange - a review, and what we have learned from STACCATO, *Journal of Geophysical Research*, *108*, 8516, 2003.

Wang, P.K., Moisture plumes above thunderstorm anvils and their contributions to cross-tropopause transport of water vapor in midlatitudes, *Journal of Geophysical Research*, *108* (D6), 4194, 2003.

Zahn, A., V. Barth, K. Pfeilsticker, and U. Platt, Deuterium, Oxygen-18, and Tritium as Tracers for Water Vapour Transport in the Lower Stratosphere and Tropopause Region, *Journal of Atmospheric Chemistry*, *30* (1), 25, 1998.

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